

Nonlocal biphoton generation in Werner state from a single semiconductor quantum dot

H. Kumano,^{1,*} H. Nakajima,¹ T. Kuroda,² T. Mano,² K. Sakoda,² and I. Suemune¹

¹*Research Institute for Electronic Science, Hokkaido University, Sapporo 001-0021, Japan*

²*National Institute for Materials Science, 1 Namiki, Tsukuba 305-0044, Japan*

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We demonstrate Werner-like polarization-entangled state generation disapproving local hidden variable theory from a single semiconductor quantum dot. By exploiting tomographic analysis with temporal gating, we find biphoton states are mapped on the Werner state, which is crucial for quantum information applications due to its versatile ramifications such as usefulness to teleportation. Observed time evolution of the biphoton state brings us systematic understanding on a relationship between tomographically reconstructed biphoton state and a set of parameters characterizing exciton state including fine-structure splitting and cross-dephasing time.

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Quantum biparticle state is the simplest physical system which could exhibit profound quantum-mechanical phenomena such as entanglement between causally independent particles [1] and nonlocality [2, 3]. These properties are the heart of quantum information and communication technology providing unconditional security [4, 5]. For two-qubit pure states, entanglement and nonlocality are equivalent [6]. In practice, however, all the systems are inevitably driven into mixed states because any system is more or less open to its environment and subject to loss and decoherence. Although the relationship between entanglement, nonlocality, and teleportation fidelity is not fully understood for general two-qubit mixed states [7, 8], one of the mixed entangled state, so-called Werner state [9, 10] has been widely investigated due to its widespread ramifications. For example, there exists bipartite mixed states which are entangled but do not violate any Bell-type inequalities [9], and all the entangled Werner states are useful for teleportation [11]. Moreover, the Werner states can be regarded as maximally entangled mixed states of two-qubit systems whose degree of entanglement cannot be increased by any unitary operations [12]. Therefore generating the Werner states is of significant importance for practical biphoton sources employed in the field of quantum information and communication technology.

For quantum photon sources with parametric down conversion [13, 14], signal and idler photons have intrinsic quantum-mechanical correlation. Biphoton states have been extensively studied with quantum state tomography [15–17], and the Werner state formation was proved by introducing polarization diffusers [18, 19]. On the other hand, quantum-dot (QD) photon sources [20], despite of their potential feasibility for quasi-deterministic operation, biphoton generation with quantum correlation is not straightforward. Lowering symmetry of exciton confinement potential results in anisotropic e - h exchange and brings fine-structure splitting (FSS) in bright exciton states [21]. Resultant which-path information hin-

ders quantum correlation by breaking superposition between two decaying paths $H_{XX}H_X$ and $V_{XX}V_X$ for neutral biexciton (XX^0)-exciton (X^0) cascading process [22]. So far, in order to suppress the which-path information for a selected QD, electric [23–25], magnetic [26], and strain [27, 28] fields, and spectral [29] or temporal [30, 31] filtering were applied, thence polarization-entangled [23–30] or nonlocal [31] photon-pair generation has been achieved. However, biphoton states generated from the QD-based sources are argued basically from a viewpoint of the state being entangled (or nonlocal) or not, and further details on the biphoton states against all the physically possible *biphoton mixed states* remain elusive yet.

In this letter, biphoton states via the XX^0 - X^0 cascading emission from a QD are systematically examined based on an analytical density matrix for the excitonic system given by Hudson et al. [32]. Density matrix which possesses the full information on the biphoton state is tomographically reconstructed and directly compared with analytically evaluated one. Highly symmetric QDs are prepared and give nonlocal biphoton mixed state without any fields or filtering process [33], which enables us to access the time evolution of the biphoton state in wider time range between XX^0 and X^0 emission. As a result, we have successfully established the quantum mechanical description for the biphoton state from a quantum dot emitter as Werner state, which distinguishes the obtained biphoton state from generally allowed biphoton mixed states. Fundamental parameters to determine underlying dynamics in the exciton state in the QD are clarified and a direction towards the ideal biphoton source is presented.

As a biphoton emitter, we employ unstrained GaAs QD formed on lattice-matched $\text{Al}_{0.3}\text{Ga}_{0.7}\text{As}$ barrier layer grown on a GaAs (111)A substrate by droplet epitaxy [34, 35]. The formed QD has typically a truncated cone shape, and average dimension of the QD is 16 nm in radius and 1.4 nm in height. Further details on the

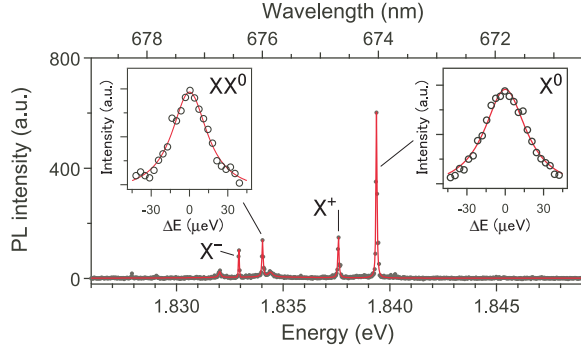


FIG. 1. PL spectrum of the isolated GaAs QD with a FSS below the system resolution of $4 \mu\text{eV}$ measured at 9K. Lorentzian fitting is also shown as a red curve. Biphoton state is generated from $|XX^0\rangle \rightarrow |X^0\rangle \rightarrow |\text{Vac}\rangle$ cascading process. Expanded spectra are also displayed for XX^0 and X^0 lines.

growth condition and the sample structure are given elsewhere [33, 34]. Since the (111) surface has C_{3v} symmetry with identical in-plane covalent bonds, one can expect mitigated QD's anisotropy [36–38], which has well verified with atomic force microscopy analysis [33]. For optical characterization, sample was cooled to 9K and a 640-nm pulsed semiconductor laser was used to pump the $\text{Al}_{0.3}\text{Ga}_{0.7}\text{As}$ barrier continuum.

Figure 1 shows the photoluminescence (PL) spectrum of an isolated GaAs dot. In our samples, QDs have typically four emission lines, i.e., negatively charged excitons (X^-), neutral biexcitons (XX^0), positively charged excitons (X^+), and neutral excitons (X^0) in order of increasing energy. For performing photon correlation measurements, we have selected the as-grown QD in which the FSS is below the system resolution of $4 \mu\text{eV}$. Biphoton correlation was investigated with a pair of cascadingly emitted XX^0 and X^0 photons by analyzing coincidence counts using time-to-digital converter (TDC). In this work, we have measured coincidence with 6^2 analyzer's polarization configurations, i.e., $\{H, V, D, A, R, L\}$ polarizations for each line. Figure 2 shows an example of accumulated coincidence counts for one analyzer configuration.

In order to analyze the biphoton state from the QD, density matrices were tomographically reconstructed from the 36 datasets entailing maximally likelihood method [15]. Degree of mixedness and entanglement are evaluated in terms of linear entropy (S_L) [39] and Tangle (T) [40], respectively. These measures can be calculated explicitly from the obtained density matrix ρ as $S_L = \frac{4}{3}(1 - \text{Tr}\rho^2)$, and $T = [\max(\lambda_1 - \lambda_2 - \lambda_3 - \lambda_4, 0)]^2$, where λ_i ($i=1, 2, 3, 4$) is the square root of the eigenvalues in decreasing order of magnitude of the spin-flipped density matrix operator $R = \rho(\sigma^y \otimes \sigma^y)\rho^*(\sigma^y \otimes \sigma^y)$, where σ^y is one of the Pauli's operators, and the asterisk indicates complex conjugation. Biphoton states can be

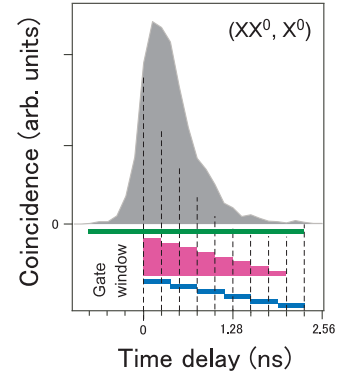


FIG. 2. (Upper panel) Typical lineshape of our histogram of the coincidence counts. Displayed data was measured with $(XX^0, X^0) = (R, L)$ configuration. (Lower panel) Integrating ranges for obtaining coincidence counts. Green stripe covers whole peak with integration time of 3.072 ns, and we refer to as *without* gating. Two types of temporal gating, (i) widening gate (red) and (ii) shifting gate (blue) are employed for evaluating the time evolution of the biphoton states generated from a highly symmetric QD. Δt_g is 256 ps and 384 ps, respectively. Time origin was determined to the point which gives the highest fidelity to $|\Phi^+\rangle$. Biphoton density matrices are tomographically reconstructed with 36 datasets for each temporal gating.

displayed in the S_L – T plane in Fig. 3(a), in which all the physically allowed biphoton states will be mapped on the white region below the dashed line. In this plane, $(S_L, T)=(0, 1)$ represents maximally entangled states, while $(S_L, T)=(1, 0)$ for totally mixed states. To begin with, we analyzed the biphoton state without temporal gating. In this case, outputs from the TDC were integrated over the range covering the whole peak, (green stripe in the lower panel of Fig. 2) and the resultant biphoton state is plotted as a green square in Fig. 3(a). The state is on the Werner curve indicated by the solid line. This is a clear manifestation that the emitted biphoton state from the QD is maximally entangled mixed state for a given S_L in the sense that none of the unitary operations will restore the entanglement further [12]. Another important consequence of this analysis is, assuming the biphoton state being the Werner state, one can readily see that the state is entangled for $S_L < 8/9$ and further violates local hidden variable model for $S_L < 1/2$ [17]. In the present case, we have $S_L=0.436$ ($< 1/2$) and $T=0.382$. We can thus conclude the generated biphoton state from the present QD is nonlocal, which is consistent with a direct demonstration of violating Clauser, Horne, Shimony, and Holt (CHSH) version of the Bell's inequality [3] without any fields nor filtering [33].

The present highly symmetric QD emitter provides biphoton states endowed with high degree of entanglement and nonlocality even without temporal gating. This achievement sheds light on the dynamics underlying the

intermediate exciton states, such as coherent evolution of the state vector and the relaxation processes involved. With a finite delay between a pair of XX^0 - X^0 photon generation and the FSS (denoted by S), possible biphoton pure state from the emitter is expressed as a bell state $|\Phi^+\rangle = \frac{1}{\sqrt{2}}(|HH\rangle + |VV\rangle)$ with a relative phase of $\exp(iSt/\hbar)$ gained in the dwell time t in the intermediate exciton state. The probability of generating the state from the QD emitter with an exciton lifetime of τ_r , at the time between t and $t + dt$, is given by $\frac{1}{\tau_r} \exp(-t/\tau_r)$. Therefore the biphoton state generated in a time duration of $[t_g, t_g + \Delta t_g]$ is

$$\hat{\rho} = \int_{t_g}^{t_g + \Delta t_g} \frac{1}{\tau_r} \exp(-t/\tau_r) \hat{\rho}_{\text{pure}}(t) dt, \quad (1)$$

where $\hat{\rho}_{\text{pure}}(t)$ is the density matrix for the pure biphoton state generated at the time t . For constructing general biphoton density matrix, we first consider effects of spin scattering and background (uncorrelated) light by taking a convex combination of the $\hat{\rho}$ and totally mixed (uncorrelated) state of $\frac{1}{4}I \otimes I$, where I indicates identity operator of a single qubit. The ratio of spin scattering (characteristic time τ_{ss}) to radiative recombination and the fraction of photon pairs stemming from the QD k define the weight of $\hat{\rho}$ as $p \equiv k/(1 + \tau_r/\tau_{ss})$. In order to describe general biphoton mixed state from the QD including relaxation processes, $1/\tau_r$ in the exponential function in Eq. 1 should be extended properly, so that population with co-polarized components ($|HH\rangle$ and $|VV\rangle$) decay with a rate of $1/\tau_r + 1/\tau_{ss}$, and decoherence takes place through further introduced parameter τ_{HV} in off-diagonal elements of the matrix to characterize cross-dephasing [32, 41]. Thus we obtain the density matrix for the biphoton mixed state with the rectilinear bases of $|HH\rangle, |HV\rangle, |VH\rangle, |VV\rangle$ as

$$\underline{\underline{\rho}} = \frac{1}{4} \begin{pmatrix} 1+p & 0 & 0 & 2pI_c^*/I_0 \\ 0 & 1-p & 0 & 0 \\ 0 & 0 & 1-p & 0 \\ 2pI_c/I_0 & 0 & 0 & 1+p \end{pmatrix}, \quad (2)$$

where

$$I_0 = \int_{t_g}^{t_g + \Delta t_g} \frac{1}{\tau_r} e^{-t(1/\tau_r + 1/\tau_{ss})} dt, \quad (3)$$

$$I_c = \int_{t_g}^{t_g + \Delta t_g} \frac{1}{\tau_r} e^{-t(1/\tau_r + 1/\tau_{ss} + 1/\tau_{HV})} e^{iSt/\hbar} dt. \quad (4)$$

If $|I_c/I_0| = 1$, hence $S = 0$ and $p' \equiv k/(1 + \tau_r/\tau_{ss} + \tau_r/\tau_{HV}) = p$ (or equivalently $\tau_r/\tau_{HV} = 0$), the biphoton state $\underline{\underline{\rho}}$ reduces to the Werner state and mapped onto an solid line in the S_L - T plane in Fig. 3(a) for $0 \leq p' \leq p \leq 1$. In order to analyze the time evolution of the biphoton states from the QD, two kind of temporal gating, i.e., (i) widening gate with constant time

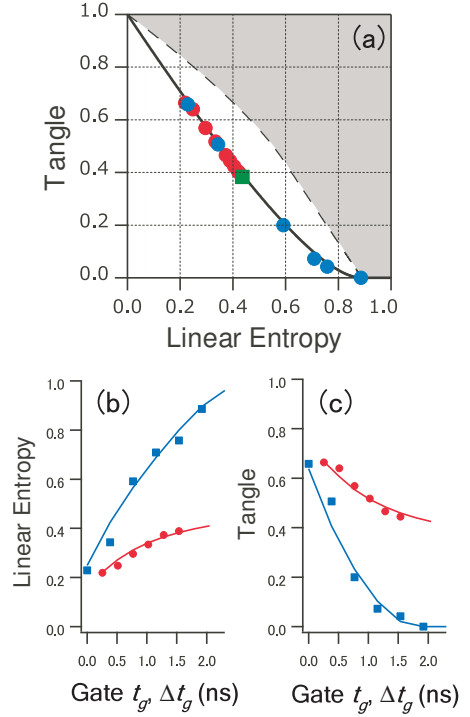


FIG. 3. Time evolution of the biphoton mixed state from a QD mapped on S_L - T plane. Werner state is denoted by solid line. (a) Experimentally obtained states via quantum tomography with 36 polarization basis are shown by red (blue) circles employing widening (shifting) gate. A green square exhibits the biphoton state without temporal gating. The corresponding time evolution of the biphoton state in terms of (b) linear entropy and (c) tangle is shown by solid lines. Horizontal axis in (b) and (c) is Δt_g (t_g) for widening (shifting) gate.

increment and (ii) shifting gate with fixed width were employed in complementary manner. Temporal gateings used for reconstructing the biphoton density matrices are illustrated in the lower panel of Fig. 2. For probing the coherent evolution, since the phase rotates as the monitoring time extends, the widening gate can be more sensitive. On the other hand, the shifting gate is preferable to evaluate the relaxation dynamics.

In Fig. 3(a), experimentally reconstructed biphoton states with the temporal gating are summarized as red (blue) circles for the widening (shifting) gate. By narrowing the gate width, the biphoton state moves toward maximally entangled state ($S_L = 0$) along the Werner curve. With the narrowest gate of 256-ps width, we have $S_L = 0.219$ and $T = 0.664$. For the shifting gate, biphoton state turned out to evolve with time along the Werner curve to the totally mixed state ($S_L = 1$) with increasing the weight of mixed state component. This finding indicates that the cross-dephasing is slow enough in comparison to the radiative lifetime ($p'/p \simeq 1$), and the phase rotation $e^{iSt/\hbar}$ contributes rather weakly to Eq. 4 ($S \simeq 0$),

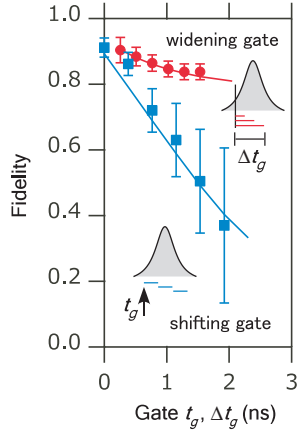


FIG. 4. Fidelity f to the $|\Phi^+\rangle = \frac{1}{\sqrt{2}}(|HH\rangle + |VV\rangle)$ for the experimentally obtained biphoton states with widening gate (red circles) and shifting gate (blue squares). Error bars represent one standard deviation. Analytically calculated fidelity employing the identical parameters with Fig. 3 is also displayed as solid lines for each gating conditions. $f \simeq (1+p)/4 + p^2/2p'$ for zero gate limit.

which suggests that $S \ll \hbar/\Delta t_g = 1.7 \mu\text{eV}$. The time evolution of the biphoton state is also displayed in more explicit way for both temporal gatings in terms of S_L and T in Fig. 3(b) and (c), respectively. In order to calculate the density matrix, the fraction of photon pairs exclusively from the QD k in $\underline{\rho}$ is required. For the analysis with temporal gating, since the k depends on the adopted gating condition, we introduce alternative parameter d to specify the ratio of uncorrelated (background) count rate to the single rate for the QD emission at zero time delay [42]. By comparing the experimentally obtained biphoton states using independently measured parameters of $\tau_r=560$ ps and $\tau_{ss}=2.8$ ns for the identical dot, we have found that $(S, \tau_{HV}, d) = (0.36 \pm 0.06 \mu\text{eV}, 2.3 \pm 0.5 \text{ ns}, 0.008 \pm 0.004)$ gives the best agreement to the experimental observation in Fig. 3(a)-(c). Basically these parameters were obtained to reproduce the upper and lower bounds for the biphoton state with shifting and widening gates in Fig. 3(a) and overall behavior in Fig. 3(b) and (c). Since the density matrix has full information on the biphoton state, we can deduce the fundamental parameters to characterize the exciton dynamics by analyzing the matrix as a function of the delay time between XX^0 and X^0 photo-generation.

In Fig. 4, entanglement fidelity f of the experimentally obtained biphoton states to the maximally entangled state $|\Phi^+\rangle$ is examined. The fidelity is calculated with $f = (1 + C_{H/V} + C_{D/A} + C_{R/L})/4$ for the two temporal gating conditions, where $C_{H/V}$, $C_{D/A}$, and $C_{R/L}$ is a correlation function in rectilinear, diagonal, and circular basis, respectively. Overall behavior agrees well with $\langle \Phi^+ | \underline{\rho} | \Phi^+ \rangle = (1 + p + 2p'\text{Re}(I_c/I_0))/4$ using the identi-

cal parameters in Fig. 3(b),(c) (solid lines), which indicates that the analytical method presented in this letter is quite useful to grasp the comprehensive understandings of the biphoton state from the QD. It is noteworthy that even for a zero gate width limit, the fidelity is below unity. This is due to residual non-zero mixedness given by $S_L \simeq (-2p^4/p'^2 - p^2 + 3)/3$. In order to realize ideal biphoton pure sources with $S_L \simeq 0$, we need $\tau_r/\tau_{ss} \simeq 0$, $\tau_r/\tau_{HV} \simeq 0$, and $k \simeq 1$. Shortening of τ_r by introducing Purcell effect [43] with background-free QDs and stabilizing spin states in the intermediate $|X^0\rangle$ state will be crucial toward this direction.

In conclusion we have demonstrated "as-grown" non-local biphoton generation from a QD without applying fields or gatings. The biphoton state generated from a QD can be well described as Werner state, which discriminates the state against generally allowed biphoton mixed states. Since the Werner state is the maximally entangled mixed biphoton state, its generation from the solid-state quantum dot in deterministic manner will be quite beneficial to the versatile fields of quantum information applications. We have also found that the cross-dephasing which loses coherence between two pathways in cascading process $|XX^0\rangle \rightarrow |X^0\rangle \rightarrow |\text{Vac}\rangle$ occurs less frequently than the exciton radiative recombination.

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* kumano@es.hokudai.ac.jp

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